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Air Effluent Monitoring

Arthur H. Biermann
Linda C. Hall
Paula J. Tate

Introduction

Lawrence Livermore National Laboratory performs continuous air effluent sampling of atmospheric discharge points at several facilities. LLNL assesses air effluent emissions from facility operations to evaluate compliance with local, state, and federal regulations and to ensure that human health and the environment are protected from hazardous and radioactive air emissions.

Air Quality Laws

LLNL complies with local, state, and federal environmental air quality laws and Department of Energy (DOE) regulations. DOE Orders 5400.1, *General Environmental Protection Program*, and 5400.5, *Radiation Protection of the Public and the Environment* define standards for controlling exposures to the public from operations at DOE facilities. Subpart H of the National Emission Standards for Hazardous Air Pollutants (NESHAPs), 40 Code of Federal Regulations (CFR) 61, requires the continuous monitoring of certain discharge points and the estimation of dose to the public resulting from operations at DOE facilities. Guidance on air effluent sampling is provided in the *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (U.S. Department of Energy 1991), 40 CFR 60, and NESHAPs-cited American National Standards Institute (ANSI) standards. In general, LLNL analyzes for most constituents at levels that are far below regulatory standards in order to determine any potential environmental impacts.

Monitored Emissions

LLNL uses a variety of radioisotopes for research purposes, including uranium, transuranics, biomedical tracers, tritium, and mixed-fission products. The major radionuclide released to the atmosphere from the Livermore site is tritium. In addition to effluent sampling for tritium, a number of facilities at the Livermore site have air effluent samplers to detect the release of uranium and transuranic aerosols. The air

effluent sampling systems described in this chapter apply to stationary and point source discharges. LLNL also monitors diffuse, or nonpoint, sources to fulfill NESHAPs requirements. Sampling methods to evaluate LLNL diffuse sources are described in Chapter 5 of the Data Supplement. Summary data from these diffuse sources can be found in Chapter 5 of this volume.

Assessment of air effluent emissions and resulting dose to the public is performed by monitoring emissions and/or evaluating potential emissions. Currently, the air effluent sampling program measures only radiological emissions. LLNL has operations with nonradiological discharges; however, permits for these operations are obtained through local agencies having enforcement authority for the Clean Air Act, and monitoring of the effluent is not required. The agencies with oversight responsibility for LLNL compliance with air regulations are Environmental Protection Agency (EPA) Region IX, the Bay Area Air Quality Management District (BAAQMD) for the Livermore site, and the San Joaquin Valley Unified Air Pollution Control District (SJVUAPCD) for Site 300. The California Air Toxics "Hot Spots" legislation requires facilities to prepare an air toxics emissions inventory and risk assessment, which LLNL has completed. Based on the assessment, the BAAQMD and the SJVUAPCD have ranked LLNL as a low-risk facility.

Historically, monitoring of radionuclide air effluents at LLNL has been implemented according to the DOE as low as reasonably achievable (ALARA) policy. This policy is meant to ensure that DOE facilities are capable of monitoring routine and nonroutine radiological releases, so that the dose to members of the public can be assessed, and so that doses are ALARA. In addition, the NESHAPs 40 CFR 61, Subpart H regulations require that facility radiological air effluents must be continuously monitored if the potential off-site dose equivalent is greater than 1 μSv/y (0.1 mrem/y), as calculated using the EPA-mandated air dispersion dose model and assuming that there are no emission control devices. The results from monitoring the air discharge points provide the actual emission source information for modeling, which is used to ensure that the NESHAPs standard, 100 μSv/y (10 mrem/y) total site effective dose equivalent, is not exceeded. Discharges from operations that have the potential to release radionuclides but that are not monitored are also evaluated according to the NESHAPs regulations, and the corresponding doses are added to those obtained by modeling monitored emissions to determine radiological NESHAPs compliance.

Operation of Monitoring Systems

Air effluent monitoring of atmospheric discharge points is used to determine the actual radionuclide releases from individual facilities and processes during routine and nonroutine operations, to confirm the operation of facility emission control systems, and to corroborate and aid in the resolution of air surveillance measurement results for the site. (The relationship can also work the other way as well—air surveillance measurements can corroborate effluent monitoring.) Measurements made by the air surveillance samplers located on and off site are reported in Chapter 5.

Methods

Air effluent monitoring involves the extraction of a measured volume of air from the exhaust of a facility or process and subsequent collection of particles by filters or of vapors by a collection medium. After collection, the various radionuclides in the sample are measured by appropriate analytical methods.

In 1998, LLNL operated 101 samplers for radioactivity from air exhausts at eight facilities at the Livermore site (see **Figure 4-1**). These systems are listed in **Table 4-1** along with the analytes of interest, the type of sampler, and the number of samplers.

LLNL reassesses the need for continuous monitoring on an annual basis and more often if warranted by new operations or changes in operations. After NESHAPs assessments of operations during 1998, LLNL installed two new continuous sampling systems to detect radioactive particles. One sampler was placed on the high-efficiency particulate air (HEPA) filtered discharge from the Extractor Test Facility in Building 177. The other sampler was installed on the HEPA-filtered exhaust from the Expedited Technology of Molten Salt Oxidation project in Building 292. Many of the other sampling systems still in place are not required by regulation; however, LLNL continues to operate these systems as a best management practice (see **Table 4-1**).

Sampling for particles containing radioactivity was conducted in seven of the facilities; sampling for tritium was conducted in one facility (Building 331). All sampling systems operated continuously. Samples were collected weekly or biweekly depending on the facility. Most air samples for particulate emissions were extracted downstream of HEPA filters and before the emissions were discharged to the atmosphere. Particles in the extracted air were collected on sample filters and analyzed for gross alpha and beta activity. Tritium is collected using molecular sieves. In addition to sample collection for environmental reporting, some facilities used real-time alarm monitors (also listed in **Table 4-1**) at discharge points to provide faster notification in the event of a release of

radioactivity. If the data are available, analytical results from the continuous samplers are reported as a measured concentration per volume of air, or as less than the minimum detection concentration (MDC) when no activity is detected. In all cases, the MDC is more than adequate for demonstrating compliance with the pertinent regulatory requirements for radionuclides that are present or may be present in the sampled air. Further details of LLNL air effluent sampling systems are included in Chapter 4 of the *Environmental Monitoring Plan* (Tate et al. 1995).

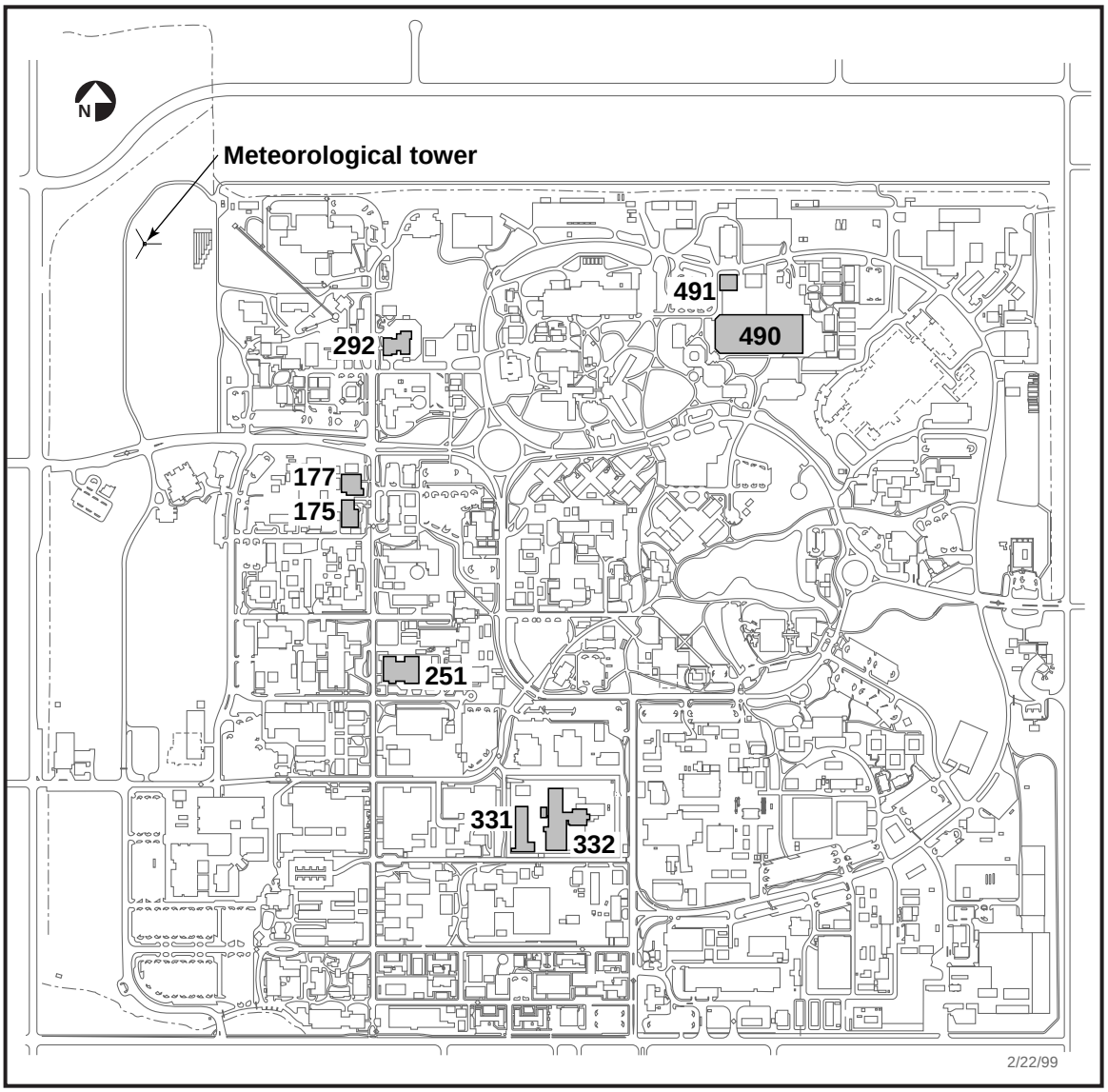

Figure 4-1. Buildings at the Livermore site with air monitoring systems for effluent gas streams during all or part of 1998.

Table 4-1. Air effluent sampling locations and systems.

Building	Facility	Analytes	Sampler type	Number of samplers
175	MARS	Gross α, β on particles	Filter	6
177	Extractor Test Facility	Gross α, β on particles	Filter	1
251	Heavy elements			
	Unhardened area	Gross α, β on particles	Filter	44
	Hardened area	Gross α, β on particles	Filter	4
	Hardened area	Gross α, β on particles	CAM[a]	4
292	Molten salt oxidation	Gross α, β on particles	Filter	1
331	Tritium	Tritium	Ionization chamber[b]	4
		Gaseous tritium and tritiated water vapor	Molecular sieves	4
332	Plutonium	Gross α, β on particles	CAM[a]	12
		Gross α, β on particles	Filter	16
490	Laser isotope separation	Gross α, β on particles	Filter	4
491	Laser isotope separation	Gross α, β on particles	Filter	1

a CAM = Eberline continuous air monitors.

b Alarmed systems.

Measured Radioactive Air Emissions

This section discusses the radiological air emissions from facilities that have continuously monitored discharge points.

Livermore Site

In 1998, a total of 4.1×10^{12} Bq (110 Ci) of tritium was released from the Tritium Facility (Building 331). Of this, approximately 3.1×10^{12} Bq (85 Ci) were released as tritiated water vapor (HTO). The remaining tritium released, 9.3×10^{11} Bq (25 Ci), was elemental tritium gas (HT). HTO emissions from the facility ranged from 220 Bq/m^3 (5.9×10^{-9} Ci/m^3) to 2.0×10^4 Bq/m^3 (5.4×10^{-7} Ci/m^3), while HT emissions ranged from 19 Bq/m^3 (5.1×10^{-10} Ci/m^3) to 3.5×10^4 Bq/m^3 (9.5×10^{-7} Ci/m^3). The highest single weekly stack emission from the facility was 3.2×10^{11} Bq (8.7 Ci), of which 5.8×10^{10} Bq (1.6 Ci) was HTO. Building 331 emissions continued to remain considerably lower than during the 1980s. **Figure 4-2** illustrates the HTO and HT emissions from the facility since

1981. In 1998, emissions from Building 331 accounted for 72% of the estimated potential tritium emissions from the Livermore site.

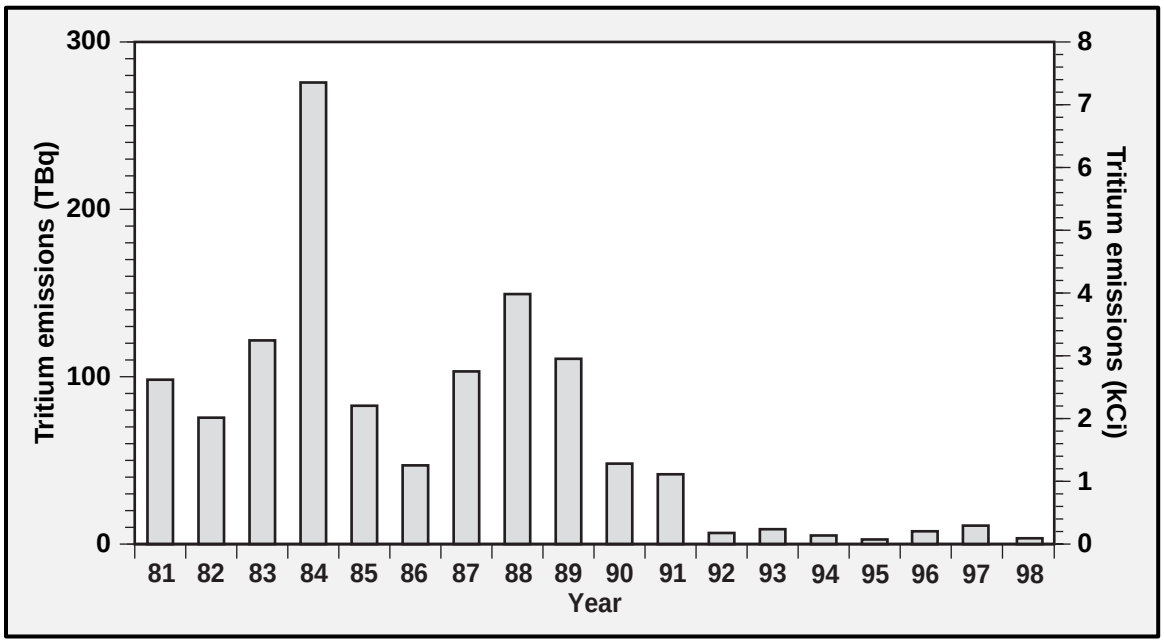

Figure 4-2. Tritium Facility HTO and HT emissions between 1981 and 1998.

Most sample results from the continuously sampled discharge points that have the potential for releasing particulate radionuclides were below the MDC of the analysis. Sometimes as few as one to four samples (out of 25 to 50 samples per year) exhibited concentrations greater than the MDC. Generally, these few samples with results above the MDC were only marginally above the MDC. In addition, because of the way some exhaust systems were configured, the monitoring systems sometimes sampled air from the ambient atmosphere and HEPA-filtered air from facility operations, which means that background atmospheric radioactivity was also collected. LLNL uses zero values for these results based on knowledge of the facility, the use of HEPA filters in all significant release pathways, and alpha-spectroscopy-based isotopic analyses of selected air sampling filters. These analyses demonstrate the presence of naturally occurring radionuclides, such as radon daughters, like polonium. Even if LLNL used the MDC values to calculate the emission estimates for these facilities (which would be an extremely conservative approach) the total dose to a member of the public attributable to LLNL activities would not be significantly affected.

In 1998, a significant number of the samples collected throughout the year from four emission points at Building 251 (the unhardened area) yielded gross alpha results greater than the MDC. We use gross alpha as the primary indicator of potential emissions for operations, such as those at Building 251 that involve the use of uranium and transuranic materials. We use gross beta results to further corroborate those gross alpha results with concentrations above the MDC. The gross alpha and gross beta activity emissions for Building 251 were 2.7×10^{3} Bq / y (7.3×10^{-8} Ci / y) and 2.3×10^{4} Bq / y (6.1×10^{-7} Ci / y). Because of the number of samples with values above the MDC, we have taken a conservative approach and are reporting gross alpha and gross beta measurements as actual emissions.

The gross alpha monitoring concentrations for Building 251 ranged from -3.0×10^{-4} Bq / m^3 (-8.2×10^{-15} Ci / m^3) to 1.4×10^{-3} Bq / m^3 (3.7×10^{-14} Ci / m^3). The Building 251 facility was in a standby, limited mode of operation, so emissions were not anticipated. It is likely that Building 251 measurements were caused by naturally occurring or background radioactivity and by the facility exhaust configuration as previously mentioned. In either case, the gross alpha and gross beta from all Livermore site operations did not a significantly contribute to the radiological dose to the public.

Table 4-2 lists total radiological emissions as determined from the continuous sampling of facility exhausts for 1998. Radioactive effluent concentrations from individual discharge points at all monitored facilities are reported in Chapter 4, Data Supplement.

Table 4-2. Measured radiological air effluent emissions for the Livermore site, 1998.

Tritium			
Building	**Facility**	**Elemental, HT (Bq)**	**Tritiated water, HTO (Bq)**
331	Tritium	9.3×10^{11}	3.1×10^{12}
Gross alpha and gross beta			
Building	**Facility**	**Gross alpha (Bq)**	**Gross beta (Bq)**
251	Heavy element	2.7×10^{3}	2.3×10^{4}

Site 300

Currently, there is no requirement for air effluent monitoring of facilities at Site 300. Air surveillance monitoring is performed for Site 300, and results are reported in Chapter 5 (Air Monitoring).

All Potential Sources of Radioactive Air Emissions

This section discusses the evaluation of all sources of radionuclide emissions to air at the Livermore site and Site 300. LLNL evaluates all discharge points with the potential to release radionuclides to the air according to 40 CFR 61, Subpart H of the NESHAPs regulations. LLNL uses radionuclide usage inventories and/or monitoring data, along with EPA-accepted release factors for operations and EPA-suggested reduction factors for emission control devices, to estimate the potential release for each individual discharge point. Potential emissions are calculated using radionuclide usage inventories as distinguished from emissions-based air effluent sampling. We conduct this evaluation annually to assess the potential dose to the public from all LLNL operations and the need for continuous sampling of individual discharge points.

In 1998, LLNL evaluated potential emissions of radionuclides from approximately 50 facilities to determine their contribution of dose to a member of the public. Potential emissions were estimated based on radionuclide usage inventories specific to individual discharge points, physical state of the materials involved in the processes, and reductions due to emission control systems. The effective dose equivalent (EDE) to a member of the public from specific operations at the Livermore site and Site 300 were published in *LLNL NESHAPs 1998 Annual Report* (Biermann et al. 1999a) and are summarized in Chapter 13 (Radiological Dose Assessment) of this report.

The radionuclide isotope responsible for the majority of the 1998 EDE was tritium. Emissions from the Tritium Facility in the form of HTO accounted for 47% of the potential EDE to the maximally exposed member of the public from the Livermore site (see Chapter 13). The dose from tritium gas is approximately 25,000 times lower than the dose from a comparable release of tritiated water; therefore, the tritiated hydrogen gas emissions did not contribute significantly to the overall tritium dose. The other measured emissions shown in **Table 4-2** (Building 251) contributed negligibly to the EDE for the maximally exposed member of the public.

When determining if continuous sampling is needed at a discharge point, LLNL evaluates operations to determine if the potential dose to the maximally exposed member of the public will exceed 0.1 mrem for the calendar year. This evaluation is similar to the evaluation of EDE previously described except no credit is allowed for emission control systems (according to the regulations). In 1998, LLNL evaluated approximately 60 potential discharge points and/or discharges at the Livermore site and Site 300. As a result of the evaluation, LLNL determined that the discharge points at Buildings 177 and 292 were the only additional discharge points that required continuous sampling.

Nonradioactive Air Emissions

The Livermore site currently emits approximately 100 kg / day of criteria air pollutants (e.g., nitrogen oxides, sulfur oxides, particulate matter [PM-10], carbon monoxide, and lead, as defined by the Clean Air Act). The largest sources of criteria pollutants from the Livermore site are surface-coating operations, internal combustion engines, solvent operations, and, when grouped together, boilers (oil and natural gas fired). **Table 4-3** lists the estimated Livermore site 1998 total airborne releases for criteria pollutants.

Table 4-3. Nonradioactive air emissions, Livermore site and Site 300, 1998.

Pollutant	Estimated releases (kg/day)	
	Livermore site	Site 300
Organics/volatile organics	25	0.90
Oxides of nitrogen	56	2.1
Carbon monoxide	11	0.48
Particulates (PM-10)	5.7	0.53
Oxides of sulfur	0.72	0.15

When comparing the estimated releases from exempt and permitted sources of air pollutants at the Livermore site to daily releases of air pollutants for the entire Bay Area, LLNL emissions are very low. For example, the total emissions of oxides of nitrogen released in the Bay Area for 1997 was approximately 1.0×10^5 kg / day compared to an estimate for LLNL releases of 56 kg / day for the Livermore site (0.056% of total Bay Area emissions). The BAAQMD estimate for reactive organic emissions was 1.2×10^5 kg / day, versus Livermore site's estimated releases of 25 kg / day (0.02% of total Bay Area emissions) in 1998.

Certain operations at Site 300 require permits from SJVUAPCD. The total estimated air emissions during 1998 from operations (permitted and exempt air sources) at Site 300 are given in **Table 4-3**. The largest sources of criteria pollutants at Site 300 include internal combustion engines, boilers, a gasoline-dispensing operation, open burning, paint spray booths, drying ovens, and soil vapor extraction operations.

Environmental Impact

Measured radiological air emissions from the Livermore site operations for 1998 are well below levels that should cause concern for public health according to existing regulatory standards for radioactive dose. The dose to the hypothetical maximally exposed member of the public due to the measured air emissions reported here (that is, due to emissions from monitored stacks) is 0.23 μSv/y (0.023 mrem/y), far below the NESHAPs standard of 100 μSv/y (10 mrem/y) and doses from naturally occurring radiation. Thus, the estimated radiological dose caused by measured air emissions from LLNL operations is minimal. See **Table 13-2** in Chapter 13 for a summary of all doses, monitored or otherwise. Nonradioactive air effluents, which are also very small compared to emissions in surrounding areas, are well below standards and are not a threat to the environment or public health.